

## Remarks

Claim 1 has been amended to more distinctly claim the present invention, including a range for the amount of synthetic cracking component in the composition and a range for unit cell size of the zeolite Y. Support for the limitation concerning synthetic cracking component may be found on page 4, lines 21-22 of the instant specification, and for the unit cell size on page 10, lines 7-8.

The essence of the present invention is a carrier composition having very specific and narrowly defined ranges of amounts of ingredients, including synthetic cracking component, as well as zeolite Y having a narrow and distinct range of unit cell size. This unique combination of ingredients achieves a surprisingly high selectivity in converting a heavy hydrocarbon feedstock into middle distillates over a broad range of conversion levels. Although the above mentioned ranges overlap much broader ranges disclosed in prior art references, such overlap does not preclude patentability of the instant claims.

It is clear from case law that such overlap does not comprise the end all to a determination of anticipation.. In *Ex parte Thumm*, 132 USPQ 66 (Board of Appeals, 1960), the claims of particular interest were rejected as lacking novelty over the prior art (Cox), i.e. a rejection under 35 USC § 102. In reversing the examiner, the Board of Appeals focused on the percentage of ethylene diamine in appellant's method as the critical feature which served to distinguish such method from the disclosure of Cox. The Cox disclosure covered a rather broad range of th diamine which in fact overlapped the more limited claimed range. The Board held that such overlap did not preclude the grant of a patent when the applicant satisfactorily establishes that he obtains results which are unobvious and unexpected and that his claims do not read upon a particular embodiment of the reference.

In *In re Wertheim*, 191 USPQ 90 (CCPA 1976), one critical parameter of the claims was a particle size of "at least 0.25 mm" and a prior art reference taught a size

of  $\frac{3}{4}$  inch which is, of course, at least 0.25 mm. The Court accurately stated that the disclosure in the prior art of any value within a claimed range is an anticipation of the claimed range, but went on to say that "to the effect that ranges which overlap or lie inside ranges disclosed by the prior art may be patentable if the applicant can show criticality in the claimed range by evidence of unexpected results".

#### Rejections Under 35 USC § 102

DeBoer et al. (WO 96/07477) is directed to a catalyst comprising a hydrogenation metal component and a clay with specified properties. It mentions as one example a catalyst composition comprising 3-40 wt.% of hydrogenation metals, 1-60 wt.% of clay component, preferably 10-50 wt.%, and 3-55 wt.% of zeolite, preferably 10-50 wt.%, and the balance alumina (paragraph bridging pages 15 and 16). On page 16, first whole paragraph, a catalyst which comprises a clay component and a zeolite Y with a unit cell size of 24.00-24.80 Å. is described as suitable for the production of diesel by hydrocracking. None of the examples describe zeolite-containing catalysts.

It is submitted that the present claims are novel over DeBoer et al. because DeBoer et al. does not disclose the specific combination of a zeolite Y with a unit cell size of 24.00 to 24.35 Å in combination with the specified ranges for the amounts of zeolite and amorphous cracking component.

More in particular, the general range for the unit cell size of the zeolite Y given in DeBoer et al. is 2.400-2.480 nm. In the present claim it is required that the unit cell size be between 2.420 and 2.435 nm. This selection is in itself novel over the disclosure of DeBoer et al.

Additionally, the amount of zeolite in the catalyst of DeBoer et al., if present at all, is specified to be 3-55 wt%, based on the total weight of the catalyst, preferably 10-50 wt.%. This corresponds to 3-92 wt%, based on the total weight of the carrier

composition, preferably 10-83%, if 3-40 wt% of hydrogenation metal is present in the catalyst. In the present claims, the amount of zeolite is selected to be 1-25 wt.%, calculated on the carrier composition. This is a selection of a small range as compared to the preferred range of 10-83% indicated in DeBoer et al.

The amount of clay component specified in DeBoer et al. is 1-60 wt.%, preferably 10-50 wt.%, calculated on the weight of the catalyst. This corresponds to 1-100 wt%, based on the total weight of the carrier composition, preferably 10-83%, if 3-40 wt% of hydrogenation metal is present in the catalyst. The present claim requires a specified amount of synthetic cracking component of at least 40 wt.%. Again, this is a selection within the broad disclosure of DeBoer et al.

Thus, as compared to the disclosure of DeBoer et al., the present claims require a selection of a specific amount of zeolite with a specific unit cell size in combination with a specific amount of synthetic cracking component. This specific combination of features is not disclosed and so is a novel selection over the general disclosure of DeBoer et al. Therefore, claim 1, and all claims dependent thereon, should be considered novel over DeBoer et al.

Nieman et al. (WO 00/00286) is no more relevant to the instant claims than DeBoer et al. Nieman et al and DeBoer share much of the same disclosure, including 1-60 wt% of the cogel component, 3-55wt% of zeolite and Y zeolite having a unit cell size in the range of 2.400-2.48nm. For reasons given above, Nieman et al is therefore no more relevant to the patentability of the present invention than DeBoer et al.

#### Rejections Under 35 USC § 103

The prior art cited by the examiner does not even create a *prima facie* case of obviousness, because no matter how the references (including Meyer ( US 3,951,864)) are combined one of ordinary skill in the art would still have no hint to the very critical combination of narrow ranges of parameters, as discussed above. However, applicants

still would be able to successfully rebut a *prima facie* case of obviousness, even if it was presented.

The catalyst which meets the specified requirements of the present claim as to amount of zeolite, unit cell size of the zeolite, and amount of synthetic cracking component also shows specific advantages, viz. a high activity in combination with a high selectivity, in the production of diesel from hydrocarbon feeds.

Comparative data is submitted in the examples of the instant application between two catalysts of the invention and a comparative catalyst. The two catalysts according to the invention comprise the synthetic cracking component specified in the present claims, viz. 60 wt.% of a cogel or a saponite clay mineral (calculated on the carrier), in combination with 10 wt.%, calculated on the carrier, of a zeolite with a unit cell size of 24.30 Å. The comparative catalyst has the same composition, except that a conventional silica-alumina cracking component is used instead of the cogel or clay mineral. From Example 4 it can be concluded that the catalysts according to the invention show a higher diesel selectivity at the same or lower ROT (operating temperature required to obtain a conversion of 70%).

Additionally, attached hereto is a Declaration of Dr. Jan Nieman, providing further experimental evidence supporting the inventiveness of the present invention.

In Example 1 of the enclosed Declaration a catalyst according to the invention based on a carrier comprising 10 wt.% of zeolite, 60 wt.% of synthetic cracking component, and the balance alumina is compared with a comparable catalyst which did not contain zeolite. It can be seen that the addition of zeolite results in a 25°C decrease of the operating temperature required to obtain 70% of conversion. This clearly is an advantageous effect.

In Example 2 of the Declaration, comparative test results are given between a catalyst according to the invention, which contains 10 wt.% of a zeolite Y with a unit cell size of 24.29 Å and a comparative catalyst which contains 10 wt.% of a zeolite Y with a unit cell size of 24.43 Å. As can be seen from the graph in this example, the catalyst according to the invention shows a surprisingly higher selectivity for diesel than the comparative catalyst at all conversion levels.

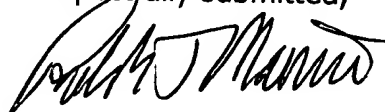
It is submitted that the experimental results in the present application and in the attached Declaration provide ample evidence to support the fact that the catalyst according to the invention with its specific requirements as to the nature of the synthetic cracking component, the amount of zeolite, and the unit cell size of the zeolite is an advantageous selection over the broad disclosure of the prior art. Therewith it is submitted that the present invention is not obvious over the prior art references cited by the Examiner, either individually, or in combination.

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#### Conclusion

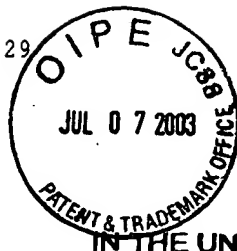
In view of the claim amendments and above remarks, it is clear that applicants have obviated basis for rejection of the instant claims, either as being anticipated by prior art references, or as being obvious over them. Allowance of the instant claims is respectfully requested.

Respectfully submitted,



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## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re                      Application of  
                             Jan Nieman et al.  
Serial No.:            09/773,381  
Filed:                  January 31, 2001

Group Art Unit:    1725  
Examiner:            C.A. Ildebrando  
Docket No.:          ACH 2850 P105

For: CATALYST CARRIER WITH HIGH DIESEL SELECTIVITY

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**Declaration under 37 C.F.R. §1.132**

Sir:

I, Jan Nieman, a citizen of the Netherlands, hereby declare and state:

1. I have a Ph.D. in chemistry, which was conferred upon me by the University of Groningen in Groningen in 1983.
2. I have been employed by Akzo Nobel since 1984 and I have a total of 19 years of work and research experience in the field of refinery catalysts.
3. I am an inventor of the present application.

I and/or those under my direct supervision and control have conducted the following tests:

Test 1: comparison of zeolite-containing catalyst according to the invention with zeolite-free catalyst

A catalyst according to the invention comprising nickel and tungsten on a carrier comprising 10 wt.% of zeolite with a unit cell size 24.30 Å, 80 wt.% of amorphous cogel and the balance alumina was prepared in accordance with Example 1 of WO 00/07723.

A comparative catalyst was prepared which did not contain the zeolite Y.

The catalysts were tested in an upflow tubular reactor. Each reactor tube contained 75 ml of catalyst homogeneously intermixed with 70 ml of carborundum particles. Before being tested the catalysts were presulfided by heating for 4 hours in a 10 vol% H<sub>2</sub>S / 90 vol% H<sub>2</sub> gas stream at 385°C and atmospheric pressure. The presulfided catalysts were then tested in hydrocracking involving a hydrotreated heavy vacuum gas oil carried out in once-through operation. The hydrotreated heavy vacuum gas oil had an initial sulfur content of 280 ppm and an initial nitrogen content of 24 ppm. Before being applied in the tests, it was spiked with dimethyldisulfide and t-butylamine until a sulfur content of 2 wt% and a nitrogen content of 1000 ppm was reached. [This spiking takes place to simulate the H<sub>2</sub>S and NH<sub>3</sub> produced by a conventional hydrocracking pretreat catalyst; in a once-through operation this H<sub>2</sub>S



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and  $\text{NH}_3$  is in practice not stripped from the feed and is passed also over hydrocracking catalyst.] The further characteristics of the hydrotreated vacuum gas oil were as follows:

**Table 1: Test feed properties**

S	wt. %	0.028 / 2
N	Ppmwt	24 / 1000
Simulated distillation	wt. %	Boiling below (°C)
	0 (IBP)	103
	5	234
	10	281
	30	389
	50	419
	70	460
	90	517
	95	541
	100 (FBP)	589

The tests were performed at three different temperatures in the range of 375–400°C, applying the following test conditions:

Hydrogen pressure	120 bar
Space velocity (LHSV)	1.00 litre feed per litre catalyst per hour (1/h)
H <sub>2</sub> /oil ratio	1000 Nl/l

The required operating temperature (ROT) was determined for a conversion of 70%. The term conversion in this context refers to the conversion, in percent, of the fraction boiling above 360°C to product with a boiling point below 360°C. The results are shown in the next Table.

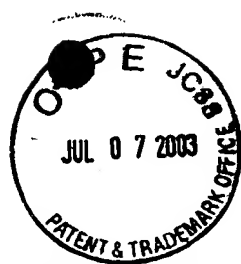
**Table 2:**

	ROT (°C)
Catalyst according to the invention	398.7
Comparative Catalyst	421.8

Table 2 shows that the catalyst according to the invention makes it possible to reduce the operating temperature of the unit with about 25°C.

**Example 2: comparison between a catalyst according to the invention and a catalyst containing a zeolite Y with a too high unit cell size**

A catalyst according to the invention comprising 6 wt.% nickel, calculated as NiO, and 21 wt.% tungsten, calculated as WO<sub>3</sub>, was prepared in accordance with Example 1 of WO 00/07723. The carrier contained 10 wt.% of zeolite Y with a unit cell size 24.29 Å, 60 wt.% of amorphous cogel and the balance alumina. In the same manner, a comparative catalyst was prepared which contained 10 wt.% of a zeolite Y with a unit cell size of 24.43 Å.



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The catalysts were tested in an upflow tubular reactor. Each reactor tube contained 100 ml of catalyst.

Before being tested the catalysts were presulfided by heating for 4 hours in a 10 vol% H<sub>2</sub>S / 90 vol% H<sub>2</sub> gas stream at 385°C and atmospheric pressure.

The presulfided catalysts were then tested in hydrocracking involving a hydrotreated heavy vacuum gas oil carried out in once-through operation. The hydrotreated heavy vacuum gas oil had an initial sulfur content of 280 ppm and an initial nitrogen content of 32 ppm. Before being applied in the tests, it was spiked with dimethyldisulfide and t-butylamine until a sulfur content of 2 wt% and a nitrogen content of 1000 ppm was reached. [This spiking takes place to simulate the H<sub>2</sub>S and NH<sub>3</sub> produced by a conventional hydrocracking pretreat catalyst; in a once-through operation this H<sub>2</sub>S and NH<sub>3</sub> is in practice not stripped from the feed and is passed also over hydrocracking catalyst.] The further characteristics of the hydrotreated vacuum gas oil were as follows:

**Table 3: Test feed properties**

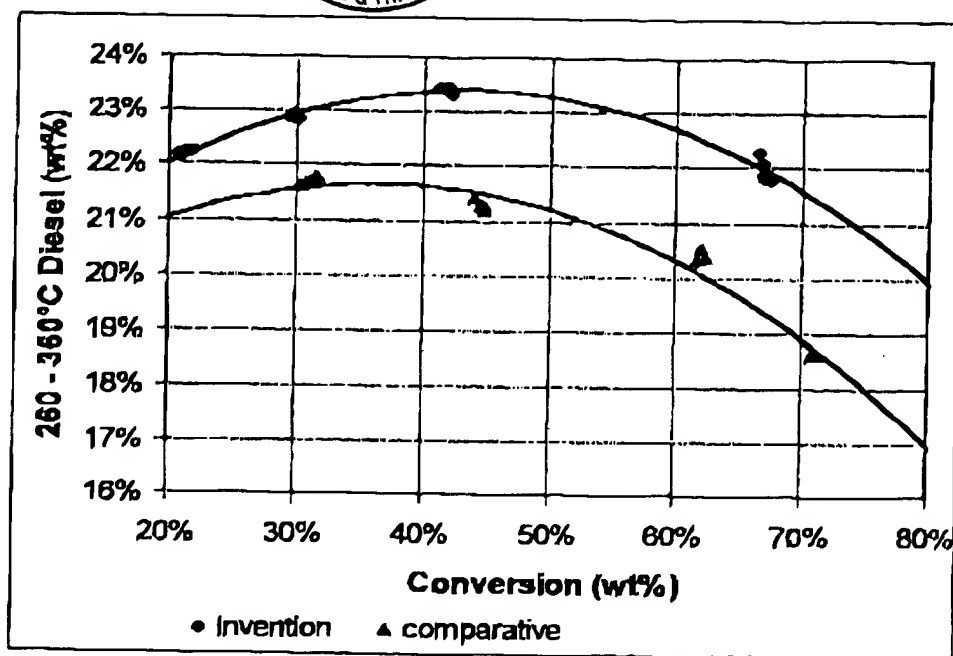
S	wt. %	0.028 / 2
N	Ppmwt	32 / 1000
Simulated distillation		
	wt. %	Bolling below (°C)
	0 (IBP)	101
	5	242
	10	289
	30	375
	50	422
	70	461
	90	517
	85	541
	100 (FBP)	585

The tests were performed at four different temperatures in the range of 368 – 401°C. applying the following test conditions:

Hydrogen pressure            120 bar  
Space velocity (LHSV)       1.00 litre feed per litre catalyst per hour (1/h)  
H<sub>2</sub>/oil ratio                    1000 NI/I

The following graph gives the diesel yield versus the conversion of the oil fraction boiling above 360°C in the feed to product boiling below 360°C.





As can be seen from the graph, the catalyst according to the invention shows a higher selectivity for diesel than the comparative catalyst at all conversion levels.

I hereby declare that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine and/or imprisonment under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing therefrom.

Date: June 27, 2003

J. Nieman